IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s):	Waheed Mukaddam *et al.*	Examiner:
Serial No.:	10/588,930	Group Art Unit:
Filing Date:	August 9, 2006	
Title:	ULTRAVIOLET LASER SYSTEM FOR DECOMPOSING CHEMICAL POLLUTANTS	

CERTIFICATE OF MAILING UNDER 37 C.F.R. § 1.8

I hereby certify that this correspondence is being deposited with the United States Post Office as First Class Mail in an envelope addressed to the Commissioner for Patents, Attention: Office of PCT Legal Administration, P.O. Box 1450, Alexandria, VA 22313-1450.

September 12, 2006
Date

Leslie R. Silverstein

Commissioner for Patents
Attention: Office of PCT Legal Administration
Attorney Cynthia M. Kratz
P.O. Box 1450
Alexandria, VA 22313-1450

PETITION TO ACCEPT APPLICATION AS FILED

Sir:

In response to the Notice of Incomplete Nonprovisional Application dated August 14, 2007, this is a Petition to accept this application as originally filed as a §111(a) application based on PCT/US2005/009500.

Reference is made to a telephone conference on August 24, 2007 between Applicants' attorney David Silverstein and Attorney Cynthia M. Kratz in the Office of PCT Legal Administration. In that telephone conference, Attorney Kratz stated that the Notice of

Incomplete Nonprovisional Application dated August 14, 2007 was apparently the result of the USPTO having misplaced the application papers that were originally filed on August 9, 2006 during the process of scanning these documents into the USPTO system. Attorney Kratz recommended that Applicants resubmit a complete set of copies of the papers filed on August 9, 2006 together with a Petition to Accept the Application as Filed. This submission is in response to that recommendation.

Accordingly, enclosed with this Petition are the following document copies:

(1) U.S. Postal Service Express Mail receipt bearing a date stamp of "Aug 09, 2006".

(2) Acknowledgement postcard as mailed; the original postcard bearing a USPTO receipt stamp was never returned.

(3) Check to "Director of the USPTO" covering application filing fees in the amount of $825.00; this check was cashed by the USPTO on August 21, 2006 (a copy of the canceled check can be provided on request), thereby clearly establishing that the USPTO received this package of materials.

(4) Certificate of Mailing certifying that the package of materials was mailed to the USPTO by Express Mail on August 9, 2006.

(5) Transmittal letter to the United States Designated/Elected Office (DO/EO/US) bearing its own Certificate of Mailing.

(6) A fully executed Declaration/Power of Attorney.

(7) A Preliminary Amendment bearing its own Certification of Mailing.

(8) An IDS bearing its own Certification of Mailing.

(9) A complete copy of the international publication WO 2005/094906 of international application PCT/US2005/009500 on which the present U.S. application is based.

As further evidence of the fact that the USPTO received the package of documents as listed above, Applicants' attorney David Silverstein notes that he had several telephone conferences in September 2006 with Supervisor Leonard Smith in the Office of PCT Legal Administration about the status of this application. Supervisor Smith confirmed that the application papers had been received and assigned Serial No. 10/588,930, but stated that this application would have to be processed as a §111(a) application because the companion application Serial No. 10/588,933 (filed the same day) was being treated as the §371 application.

Based on these discussions with Supervisor Smith, Applicants filed a Petition to Convert to §111(a) Application on September 22, 2006 and paid the Petition fee. Apparently unaware that Applicants had already filed a Petition to Convert, the USPTO sent a Notification dated 07 Nov 2006 (signed by Cynthia M. Kratz). The Notification of 07 Nov 2006 states: "On 09 August 2006, applicant filed two transmittal letters for entry into the national stage in the United States ... As is evident from the above recited facts, two sets of papers to enter the national stage were submitted...." (emphasis added).

Following telephone discussions with Attorney Kratz, a Second Preliminary Amendment for the subject application was faxed to Attorney Kratz on December 8, 2006 to identify the application as a "Continuation of International Patent Application No. PCT/US2005/009500...." Based on this further submission, Attorney Kratz issued a further Notification dated 12 Dec 2006 in which it was concluded that "...it is proper to treat the initial filing as a filing under 35 U.S.C. 111(a)."

Because all of the papers required for a §111(a) application were duly filed with the USPTO on August 9, 2006, together with the appropriate filing fee, U.S. Serial No. 10/588,930 is entitled to the August 9, 2006 filing date.

This submission is fully responsive to the Notice of Incomplete Nonprovisional Application in providing copies of the Specification as filed on August 9, 2006 and of the executed Declaration as also filed on August 9, 2006.

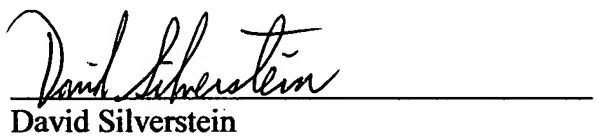
Accordingly, these papers should be forwarded to the Office of Initial Patent Examination, and a Filing Receipt for this application should promptly be issued. The Filing Receipt should preserve the August 9, 2006 filing date to which Applicants are entitled.

FEES

Because this Petition was occasioned by a USPTO oversight in misplacing the application papers, Applicants submit that no Petition or other fee should be due with this submission.

However, in the event that any fee is associated with this submission, authorization is hereby given to the USPTO to charge any such fee to Deposit Account 50-1139.

September 12, 2007
Date



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Andover, MA 01810
Tel. No. (978) 470-0990
Fax No. (978) 470-0993

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
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P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371 (c) DATE	FIRST NAMED APPLICANT	ATTORNEY DOCKET NUMBER
10/588,930	08/09/2006	Waheed Mukaddam	WLI-001A

CONFIRMATION NO. 3009

FORMALITIES LETTER

David Silverstein
Andover-IP-Law
Suite 300
44 Park Street
Andover, MA 01810

Date Mailed: 08/14/2007

NOTICE OF INCOMPLETE NONPROVISIONAL APPLICATION

FILED UNDER 37 CFR 1.53(b)

A filing date has NOT been accorded to the above-identified application papers for the reason(s) indicated below.

All of the items noted below **and a newly executed oath or declaration covering the items must** be submitted within **TWO MONTHS** of the date of this Notice, unless otherwise indicated, or proceedings on the application will be terminated *(37 CFR 1.53(e))*. Replies should be mailed to: Mail Stop Missing Parts, Commissioner for Patents, P.O. Box 1450, Alexandria VA 22313-1450.

The filing date will be the date of receipt of all items required below, unless otherwise indicated. Any assertions that the item(s) required below were submitted, or are not necessary for a filing date, must be by way of petition directed to the attention of the Office of Petitions accompanied by the $400.00 petition fee *(37 CFR 1.17(f))*. If the petition states that the application is entitled to a filing date, a request for a refund of the petition fee may be included in the petition.

If the above-identified application contains a priority claim under 37 CFR 1.55 or benefit claim under 37 CFR 1.78 of a prior-filed application that was present on the filing date of the application and applicant wants to rely on 37 CFR 1.57(a) to add inadvertently omitted material to the above-identified application, applicant must file a petition under 37 CFR 1.57(a) accompanied by the $400.00 petition fee (37 CFR 1.17(f)) within TWO MONTHS of the date of this Notice. Petitions should be mailed to: Mail Stop Petitions, Commissioner for Patents, P.O. Box 1450, Alexandria VA 22313-1450.

- The specification is missing.
  A complete specification as prescribed by 35 U.S.C. 112 is required.

Applicant is cautioned that correction of the above items may cause the specification and drawings page count to exceed 100 pages. If the specification and drawings exceed 100 pages, applicant will need to submit the required application size fee.

Replies should be mailed to: Mail Stop Missing Parts
Commissioner for Patents
P.O. Box 1450
Alexandria VA 22313-1450

Registered users of EFS-Web may alternatively submit their reply to this notice via EFS-Web.
https://sportal.uspto.gov/authenticate/AuthenticateUserLocalEPF.html

For more information about EFS-Web please call the USPTO Electronic Business Center at **1-866-217-9197** or visit our website at http://www.uspto.gov/ebc.

If you are not using EFS-Web to submit your reply, you must include a copy of this notice.

Office of Initial Patent Examination (571) 272-4000, or 1-800-PTO-9199
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Mailer Signature

FROM: (PLEASE PRINT) PHONE (978) 470-0990

WLI-001A

David Silverstein
Andover-IP-Law
44 Park Street, Suite 300
Andover, MA 01810

TO: (PLEASE PRINT) PHONE ( )

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Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

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EMS

COPY

Re Application of:
Waheed Mukaddam et al.

Due Date: Sept. 25, 2006
Mailed: Aug. 9, 2006
Mailer: DS/lrs

For: Ultraviolet Laser System for Decomposing Chemical Pollutants

Serial No. | Patent No. | Docket: WLI-001A

In the above matter, the following has been received in the U.S. Patent and Trademark Office on the date stamped hereon.

- [X] Specification, Abstract and (Sec. 371 + Cover Page) (30) Claims — (24) Total Pages
- [X] Declaration and Power of Attorney
- [X] Drawing (1 sheets) (Figs. 1 and 2)
- [ ] Assignment _____
- [X] Check $ 825.00
- [ ] Request for Recordation
- [X] Information Disclosure Statement
- [X] Small entity verification
- [X] Transmittal Letter to DO/EO/US – Filing Under 35 U.S.C. 371
- [ ] Extension of time _____
- [XX] Amendment Preliminary
- [XX] Cert. of Mailing Express Mail
- [ ] Maintenance Fee Transmittal
- [ ] Affidavit (w/wo Exhibits)
- [ ] Notice of Appeal
- [ ] Brief ( ) copies
- [ ] Issue fee transmittal
- [XX] Transmittal letter

EQ142620626US

1388

ANDOVER-IP-LAW
ATTORNEY DAVID SILVERSTEIN
44 PARK STREET, #300
ANDOVER, MA 01810

DATE August 9, 2006

5-7515/110

PAY TO THE ORDER OF Director of the USPTO $ 825.00/xx

Eight hundred twenty five and 00/xx DOLLARS

Sovereign Bank
sovereignbank.com

FOR WLI-001A — Sec. 371 Filing fee

David Silverstein

MP

⑈001388⑈ ⑆011075150⑆ 873000526821⑈

COPY

IN THE UNITED STATES PATENT OFFICE
AS DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

Applicant:	Waheed Mukaddam et al.
International Application No.:	PCT/US2005/009500
International Filing Date:	22 March 2005 (22.03.2005)
Title:	ULTRAVIOLET LASER SYSTEM FOR DECOMPOSING CHEMICAL POLLUTANTS
Agent's File Reference:	WLI-001A

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

"Express Mail" Mailing Label Number: EQ 142620626 US
Date of Deposit: August 9, 2006

I hereby certify that the attached international application PCT/US2005/009500 published as WO 2005/094906, a Transmittal Letter Under 35 U.S.C. 371, an executed Declaration/Power of Attorney, an Information Disclosure Statement, check for $825.00, and a mailroom postcard are being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 C.F.R. § 1.10 on the date indicated above and are addressed to Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

Leslie R. Silverstein
(Type or print name of person mailing paper)

(Signature of person mailing paper)

PTO-1390 (Rev. 07-2005)
Approved for use through 3/31/2007. OMB 0651-0021
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A SUBMISSION UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER	
WLI-001A	
U.S. APPLICATION NO. (If known, see 37 CFR 1.5)	
INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE
PCT/US2005/009500	22 March 2005 (22.03.2005)
PRIORITY DATE CLAIMED	
25 March 2004 (25.03.2004)	

TITLE OF INVENTION
Ultraviolet Laser System for Decomposing Chemical Pollutants

APPLICANT(S) FOR DO/EO/US
Waheed Mukaddam et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [X] This is a **FIRST** submission of items concerning a submission under 35 U.S.C. 371.
2. [ ] This is a **SECOND** or **SUBSEQUENT** submission of items concerning a submission under 35 U.S.C. 371.
3. [X] This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. [X] The US has been elected (Article 31).
5. [X] A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. [XX] is attached hereto (required only if not communicated by the International Bureau).
   b. [ ] has been communicated by the International Bureau.
   c. [XX] is not required, as the application was filed in the United States Receiving Office (RO/US).
6. [X] An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
   a. [X] is attached hereto.
   b. [ ] has been previously submitted under 35 U.S.C. 154(d)(4).
7. [X] Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. [ ] are attached hereto (required only if not communicated by the International Bureau).
   b. [ ] have been communicated by the International Bureau.
   c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
   d. [XX] have not been made and will not be made.
8. [ ] An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. [X] An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. [ ] An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. [X] An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. [ ] An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. [X] A preliminary amendment. (Substituting new claims 31 - 71 for original claims 1 - 30)
14. [ ] An Application Data Sheet under 37 CFR 1.76.
15. [ ] A substitute specification.
16. [X] A power of attorney and/or change of address letter. (included with inventor Declaration)
17. [ ] A computer-readable form of the sequence listing in accordance with PCT Rule 13*ter*.2 and 37 CFR 1.821- 1.825.
18. [ ] A second copy of the published International Application under 35 U.S.C. 154(d)(4).
19. [ ] A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).

PTO-1390 (Rev. 07-2005)
Approved for use through 3/31/2007. OMB 0651-0021
U.S. Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
	PCT/US2005/009500	WLI-001A

20. Other items or information:

The following fees have been submitted	CALCULATIONS	PTO USE ONLY
21. [X] Basic national fee (37 CFR 1.492(a)) $300	$ 300.00	
22. [X] Examination fee (37 CFR 1.492(c)) If the written opinion prepared by ISA/US or the international preliminary examination report prepared by IPEA/US indicates all claims satisfy provisions of PCT Article 33(1)-(4) ..... $0 All other situations $200	$ 200.00	
23. [X] Search fee (37 CFR 1.492(b)) If the written opinion of the ISA/US or the International preliminary examination report prepared by IPEA/US indicates all claims satisfy provisions of PCT Article 33(1)-(4) ..... $0 Search fee (37 CFR 1.445(a)(2)) has been paid on the international application to the USPTO as an International Searching Authority ..... $100 International Search Report prepared by an ISA other than the US and provided to the Office or previously communicated to the US by the IB ..... $400 All other situations $500	$ 100.00	
TOTAL OF 21, 22 and 23 =	$ 600.00	
[ ] Additional fee for specification and drawings filed in paper over 100 sheets (excluding sequence listing in compliance with 37 CFR 1.821(c) or (e) or computer program listing in an electronic medium) (37 CFR 1.492(j)). The fee is $250 for each additional 50 sheets of paper or fraction thereof.		
Total Sheets: - 100 = / Extra Sheets: /50 = / Number of each additional 50 or fraction thereof (round up to a whole number): / RATE: x $250	$	
Surcharge of $130.00 for furnishing any of the search fee, examination fee, or the oath or declaration after the date of commencement of the national stage (37 CFR 1.492(h)).	$	

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	$	
Total claims	41 - 20 =	21	x $50	$ 1,050.00	
Independent claims	2 - 3 =	0	x $200	$ -0-	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ $360	$ N/A	
			TOTAL OF ABOVE CALCULATIONS =	$ 1,650.00	
[X] Applicant claims small entity status. See 37 CFR 1.27. Fees above are reduced by ½.					
			SUBTOTAL =	$ 825.00	
Processing fee of $130.00 for furnishing the English translation later than 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +				$ N/A	
			TOTAL NATIONAL FEE =	$ 825.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). $40.00 per property +				$ N/A	
			TOTAL FEES ENCLOSED =	$ 825.00	
			Amount to be refunded:	$	
			Amount to be charged	$	

PTO-1390 (Rev. 07-2005)
Approved for use through 3/31/2007. OMB 0651-0021
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

a. [XXX] A check in the amount of $ $825.00 to cover the above fees is enclosed.

b. [ ] Please charge my Deposit Account No. _____ in the amount of $ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. [XXX] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 50-1139. A duplicate copy of this sheet is enclosed.

d. [ ] Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the International Application to pending status.

SEND ALL CORRESPONDENCE TO:

David Silverstein
Andover-IP-Law
44 Park Street, Suite 300
Andover, MA 01810

David Silverstein
SIGNATURE

David Silverstein
NAME

26,336
REGISTRATION NUMBER

EXPRESS MAILING UNDER 37 C.F.R. § 1.10*
(Express Mail label number is mandatory.)
(Express Mail certification is optional.)

I hereby certify that this paper, along with any document referred to, is being deposited with the United States Postal Service on this date August 9, 2006, in an envelope addressed to the Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 as "Express Mail Post Office to Addressee" Mailing Label No. EQ 142620626 US

Leslie R. Silverstein
(type or print name of person mailing paper)

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY
(Includes Reference to PCT International Applications)

ATTORNEY'S DOCKET NUMBER: WLI-001A

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ULTRAVIOLET LASER SYSTEM FOR DECOMPOSING CHEMICAL POLLUTANTS

the specification of which (check only one item below):

☐ is attached hereto.

☐ was filed as United States application

Serial No. _____

on _____.

and was amended

on _____ (if applicable).

☒ was filed as PCT international application

Number PCT/US2005/009500

on 22 Mar 2005 (22.03.2005).

and was amended under PCT Article 19

on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119:

COUNTRY (if PCT indicate PCT)	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 USC 119
			☐ YES ☐ NO
			☐ YES ☐ NO
			☐ YES ☐ NO
			☐ YES ☐ NO
			☐ YES ☐ NO

Combined Declaration For Patent Application and Power of Attorney (Continued)

ATTORNEY'S DOCKET NUMBER: WLI-001A

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:

PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C 120

U.S. APPLICATIONS		STATUS (Check one)		
U.S. APPLICATION NUMBER	U.S. FILING DATE	PATENTED	PENDING	ABANDONED
60/556,463	25 Mar 2004 (25.03.2004)			X

PCT APPLICATIONS DESIGNATING THE U.S.					
PCT APPLICATION NO.	PCT FILING DATE	U.S. SERIAL NUMBERS ASSIGNED (if any)			
PCT/US2005/009500	22 Mar 2005			X	
	(22.03.2005)				

POWER OF ATTORNEY. As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. *(List name and registration number)*

David Silverstein, Reg. No. 26,336

Send Correspondence to.

David Silverstein
Andover-IP-Law
44 Park Street, Suite 300
Andover, MA 01810

Direct Telephone Calls to *(name and telephone number)*

David Silverstein
(978) 470-0990

201	FULL NAME OF INVENTOR	FAMILY NAME: MUKADDAM	FIRST GIVEN NAME: Waheed	SECOND GIVEN NAME:
	RESIDENCE & CITIZENSHIP	CITY: Cambridge	STATE OR FOREIGN COUNTRY: Massachusetts	COUNTRY OF CITIZENSHIP: USA
	POST OFFICE ADDRESS	POST OFFICE ADDRESS: 72 Pleasant Street	CITY: Cambridge	STATE & ZIP CODE/COUNTRY: MA 02139 USA
202	FULL NAME OF INVENTOR	FAMILY NAME: THOMPSON	FIRST GIVEN NAME: Allan	SECOND GIVEN NAME: R.
	RESIDENCE & CITIZENSHIP	CITY: Winchester	STATE OR FOREIGN COUNTRY: Massachusetts	COUNTRY OF CITIZENSHIP: U.S.A.
	POST OFFICE ADDRESS	POST OFFICE ADDRESS: 31 Holton Street	CITY: Winchester	STATE & ZIP CODE/COUNTRY: MA 01890 U.S.A.
203	FULL NAME OF INVENTOR	FAMILY NAME: MOORES	FIRST GIVEN NAME: Charles	SECOND GIVEN NAME: W.
	RESIDENCE & CITIZENSHIP	CITY: Wayland	STATE OR FOREIGN COUNTRY: Massachusetts	COUNTRY OF CITIZENSHIP: U.S.A.
	POST OFFICE ADDRESS	POST OFFICE ADDRESS: 4 Deer Run	CITY: Wayland	STATE & ZIP CODE/COUNTRY: MA 01778 U.S.A.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon

SIGNATURE OF INVENTOR 201	SIGNATURE OF INVENTOR 202	SIGNATURE OF INVENTOR 203
✓	✓	✓
DATE: ✓ Aug. 3, 2006	DATE: ✓ August 3, 2006	DATE: ✓ August 3, 2006

PTO 1391 (Rev. 8 03) Page 2 of 2

U.S. DEPARTMENT OF COMMERCE Patent and Trademark Office

Waheed Mukaddam Allan R. Thompson Charles W. Moores

Practitioner's Docket No. WLI-001A

AMENDMENT ACCOMPANYING NEW APPLICATION TRANSMITTAL

TO THE UNITED STATES ELECTED OFFICE (EO/US)
(ENTRY INTO U.S. NATIONAL PHASE UNDER CHAPTER II)

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/US2005/009500	22 March 2005 (22.03.2005)	25 March 2004 (25.03.2004)

TITLE OF INVENTION

Ultraviolet Laser System for Decomposing Chemical Pollutants

APPLICANT(S)

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COPY

Please amend the accompanying new patent application under 35 U.S.C. Sec. 371 as follows:

Amendments to the Specification begin on page 2 of this paper.

Amendments to the Claims are reflected in the Listing of Claims which begins on page 5 of this paper.

Remarks begin on page 16 of this paper.

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Amendments to the Specification

In the Specification

Please substitute the following paragraph for the paragraph at page 5, lines 17-20:

Still another object of this invention is provide a UV-laser based water treatment system and methods to irradiate water with monochromatic laser light, all or a predominant portion of which is at wavelengths within the ultraviolet range of about 180 nm to 400nm (55,560 cm^{-1} to 25,000 cm^{-1}) and, in some preferred embodiments for selected applications, about 193 nm (51,810 cm^{-1}) or lower.

Please substitute the following paragraph for the paragraph at page 7, lines 1-7:

only a few minutes or less of exposure of a water sample containing the chemicals to UV laser radiation, preferably at one, or several, or within a range of monochromatic UV wavelengths of about 180 nm to 400nm (55,560 cm^{-1} to 25,000 cm^{-1}), in some instances preferably about 193 nm (51,810 cm^{-1}) or lower. It has been consistently found that a UV laser treatment in accordance with this invention results in higher decomposition/ destruction of chemical constituents in a treated aqueous sample over a dramatically shorter period of treatment time than does light from other photonic sources, specifically light from a UV lamp.

Please substitute the following paragraph for the paragraph at page 7, lines 8-19:

In a preferred embodiment of this invention, the UV laser radiation wavelength(s) used is (are) selected in relation to the chemical substance(s) to be decomposed in a particular aqueous sample. It has been found that UV laser radiation wavelengths or wavelength ranges can be effectively coordinated with chemical atomic bonding energies. Thus, in accordance with an embodiment of this invention, it may be possible to identify a preferred UV laser radiation wavelength for decomposing a single chemical constituent, or to identify preferred multiple UV laser radiation wavelengths, or a preferred UV laser radiation wavelength range, for decomposing multiple chemical constituents or for addressing a single chemical constituent that may decompose into different intermediate substances prior to complete decomposition. In some instances, it has been found that UV laser radiation at about 193 nm (51,810 cm^{-1}) or lower, i.e., about 180 nm – 193 nm (55,560 cm^{-1} to 51,810 cm^{-1}) is particularly effective in decomposing many of the chemical constituents of primary interest.

Please substitute the following paragraph for the paragraph at page 15, lines 8-24:

For example, in demonstrating the practice of one embodiment of this invention, a distilled water blank and various concentrations, ranging from about 5 ppm to about 500 ppm, of ammonium perfluorooctanoate (apfo) in distilled water were prepared. A portion of each apfo sample was set aside as a reference and another portion of each sample was introduced into a 22mm diameter by 50mm long synthetic quartz (Suprasil™) reaction vessel. The water solutions of apfo were irradiated by means of an excimer laser (LambdaPhysik LPX210i) producing a monochromatic 193 nm (51, 810 cm^{-1}) wavelength beam operating at an energy level of 100 millijoules per pulse at a frequency of 50 pulses

per second. The laser beam was directed along (parallel to) the long axis of the reaction vessel and completely covered an 8mm high by 22 mm wide optical window section of the reaction vessel so as to fill an 8mm by 22mm portion of the reactor cavity with UV laser light resulting in the delivery of a UV light energy intensity of about 0.57 millijoules/square millimeter/pulse to the sample. A 30w deuterium lamp and Zeiss MMS-UV spectrometer were mounted perpendicular to the long axis of the reaction vessel and approximately in the middle of the reaction vessel to measure the ultraviolet spectrum of the contents of the reaction vessel as a function of time while the sample was being exposed to the UV laser light. Various parameters from the experimental system were interfaced to a computer system electrically

Listing of Claims

Please amend the claims as follows. This Listing of Claims will replace all prior versions and listings of claims in this application:

Claims

1. – 30. (Canceled)

31. (New) A method for substantially completely decomposing undesirable inorganic chemical substance(s) in an aqueous solution and/or dispersion, said method comprising the ultraviolet laser treatment step of exposing an aqueous portion containing one or more undesirable chemical substance(s) to ultraviolet laser irradiation at a suitable ultraviolet wavelength and at a sufficient energy density for a sufficient period of time of about 15 minutes or less substantially to decompose the undesirable chemical substance(s) in the aqueous portion.

32. (New) A method according to claim 31 wherein the ultraviolet laser irradiation used for the ultraviolet laser treatment step consists substantially exclusively of ultraviolet laser irradiation having a wavelength of about 180 nm to 400 nm (55,560 cm^{-1} to 25,000 cm^{-1}) that delivers an energy density in the range of about 0.10 to 10 millijoules per square millimeter of ultraviolet laser beam cross-section to said aqueous portion.

33. (New) A method according to claim 31 wherein the ultraviolet laser irradiation consists substantially exclusively of ultraviolet laser irradiation having a wavelength of about 193 nm (51,810 cm^{-1}) or lower.

34. (New) A method according to claim 31 wherein the total period of time during which the aqueous portion is exposed to the ultraviolet laser irradiation to achieve substantially complete decomposition of at least one of the undesirable chemical substances is about 1 second to 10 minutes.

35. (New) A method according to claim 31 wherein the total period of time during which the aqueous portion is exposed to the ultraviolet laser irradiation to achieve substantially complete decomposition of at least one of the undesirable chemical substances is less than one second.

36. (New) A method according to claim 31 wherein 90% or more of at least one chemical substance originally present in the aqueous portion is decomposed during an ultraviolet laser irradiation period of less than 15 minutes.

37. (New) A method according to claim 31 wherein the aqueous portion, prior to the ultraviolet laser treatment step, contains one or more perchlorate compounds and, after said ultraviolet laser treatment step, the aqueous portion contains substantially no perchlorate compounds.

38. (New) A method according to claim 31 wherein a catalyst is added to the aqueous portion before or during the ultraviolet laser irradiation step.

39. (New) A method according to claim 31 wherein said ultraviolet laser irradiation is delivered to the aqueous portion in pulses at a pulse rate ranging from about 1 to 50,000 pulses per second.

40. (New) A method according to claim 31 wherein said ultraviolet laser irradiation is delivered to the aqueous portion in pulses at a pulse rate ranging from about 10 to 1000 pulses per second.

41. (New) A method according to claim 31 wherein said ultraviolet laser irradiation is delivered to the aqueous portion in pulses at a pulse rate ranging from about 25 to 100 pulses per second.

42. (New) A method according to claim 31 further comprising the step of applying a combination of an ultraviolet laser pulse rate ranging from about 10 to 1000 pulses per second and an energy density ranging from about 0.10 to 10 millijoules per square millimeter of ultraviolet laser beam cross-section so as to deliver to the aqueous portion sufficient laser energy to effect substantially complete decomposition of said undesirable chemical substance(s) within a total treatment time of about 15 minutes or less.

43. (New) A method according to claim 31 further comprising the step of applying a combination of an ultraviolet laser wavelength or wavelengths within the range of about 180 nm to about 400 nm (55,560 cm^{-1} to 25,000 cm^{-1}) an ultraviolet laser pulse rate ranging from about 10 to 1000 pulses per second, and an energy density ranging from about 0.10 to 10 millijoules per square millimeter of ultraviolet laser beam cross-section so as to deliver to the aqueous portion sufficient laser energy at suitable wavelengths to effect substantially complete decomposition of said chemical substance(s) within a total treatment time of about 15 minutes or less.

44. (New) A method according to claim 31 further comprising a monitoring step of periodically or continuously monitoring the concentration of the undesirable chemical substances in the aqueous portion during the ultraviolet laser treatment step.

45. (New) A method according to claim 44 wherein said aqueous portion is treated with an ultraviolet laser light beam while flowing in a conduit having a longitudinal axis, said laser light beam being oriented substantially parallel to the longitudinal axis of said conduit, wherein said monitoring step is performed continuously during the ultraviolet laser treatment step at two or more locations along the axis of said conduit.

46. (New) A method according to claim 44 wherein said monitoring step comprises the steps of passing a light beam: (a) through a first optically transparent wall portion of a container holding said aqueous portion during the ultraviolet laser treatment step, (b) through the aqueous portion being treated with ultraviolet laser radiation in said container,

(c) out of said container through a second optically transparent wall portion of said container, and (d) into a spectrometer for monitoring the spectrophotometric signature of the aqueous portion during the ultraviolet laser treatment step.

47. (New) A method according to claim 31 wherein the aqueous portion is exposed to the ultraviolet laser irradiation in a batch, semi-batch, or continuous flow process.

48. (New) A method according to claim 31 wherein the treatment step of exposing the aqueous portion containing undesirable chemical substance(s) to ultraviolet laser irradiation is carried out without adding an oxidant to the aqueous portion.

49. (New) A method according to claim 31 wherein the treatment step reduces the concentration of the chemical substance(s) in the aqueous portion to a level that is consistent with environmental regulations for discharging a treated aqueous portion to the environment.

50. (New) A method according to claim 31 wherein the ultraviolet laser irradiation delivers at least ten times the energy of a conventional ultraviolet lamp light beam per square millimeter of ultraviolet laser beam cross-section to the aqueous portion.

51. (New) A method according to claim 31 wherein the ultraviolet laser irradiation consists of highly monochromatic ultraviolet laser light at one or several distinct monochromatic ultraviolet wavelengths.

52. (New) A method according to claim 51 further comprising the steps of determining the one or several monochromatic ultraviolet laser light wavelengths which are the most effective ultraviolet laser light wavelengths for decomposing the undesirable chemical substance(s) present in the aqueous portion, and delivering to the aqueous portion ultraviolet laser irradiation substantially consisting of only such most effective ultraviolet laser light wavelengths.

53. (New) A method according to claim 31 further comprising the steps of determining the one or several ultraviolet laser light wavelengths which are the most effective ultraviolet laser light wavelengths in decomposing the undesirable chemical substance(s) present in the aqueous portion based on the respective chemical atomic bonding energies of those undesirable chemical substance(s), and delivering to the aqueous portion ultraviolet laser irradiation substantially consisting of only such most effective ultraviolet laser light wavelengths.

54. (New) A method according to claim 31 further comprising the steps of: directing an ultraviolet laser beam from an ultraviolet laser device into a reaction vessel containing the aqueous portion through a substantially ultraviolet-transparent optical window portion so as to irradiate the contents of the reaction vessel; orienting a deuterium lamp toward the reaction vessel so as to direct light beams through the reaction vessel substantially at right angles to the ultraviolet laser beam; and, continuously monitoring the ultraviolet spectrophotometric signature of the contents of the reaction vessel using an ultraviolet spectrophotometric system to provide an indication of the chemical decomposition reactions occurring within the reaction vessel.

55. (New) A method according to claim 54 further comprising the step of turning off the laser and removing the treated aqueous portion when the rate of change of the spectrophotometric signature significantly slows or ceases and thereby indicates substantial completion of the chemical decomposition process.

56. (New) A method according to claim 55 wherein a water sample containing a known concentration of the undesirable chemical substances(s) of interest is placed in the reaction vessel and is treated by the ultraviolet laser treatment, the irradiated water sample is thereafter removed for further analysis, and the results of such analysis are used to ascertain the completeness of the chemical decomposition of the undesirable chemical substance(s).

57. (New) A method according to claim 56 wherein water samples with varying known concentrations of the undesirable chemical substance(s) are irradiated with ultraviolet laser irradiation according to the method, and the results are compared for effectiveness in decomposing the substance(s).

58. (New) A method according to claim 31 further comprising the steps of: preparing test samples consisting of a distilled water blank and different aqueous perchlorate solutions containing from 5 ppm to 500 ppm of perchlorate in distilled water; setting aside one portion of each perchlorate solution as a reference portion; treating another portion of each perchlorate solution in a synthetic quartz reaction vessel by irradiating each sample with an excimer laser producing a monochromatic 193 nm (51,810 cm^{-1}) wavelength beam

operating at an energy level of 100 millijoules per pulse at a frequency of 50 pulses per second, wherein the laser beam is directed along the long axis of the reaction vessel and completely covers an optical window section of the reaction vessel so as to substantially fill the reactor cavity with ultraviolet laser light and to deliver an ultraviolet laser light energy intensity of about 0.57 millijoules/square millimeter/pulse, based on the area of the optical window section, to each treated perchlorate portion thereby obtaining 95% or greater decomposition of the perchlorate in each treated perchlorate portion over a period of less than 15 minutes of irradiation.

59. (New) A method according to claim 58 further comprising the steps of: mounting a deuterium lamp and a spectrometer perpendicular to the long axis of the reaction vessel; directing light from the deuterium lamp through the reaction vessel and the contents of the reaction vessel and thereafter into the spectrometer; and measuring the ultraviolet spectrum of the contents of the reaction vessel as a function of time while each sample is being exposed to the ultraviolet laser light.

60. (New) Apparatus for treating an aqueous solution and/or dispersion of undesirable inorganic chemical substance(s) so as to substantially completely decompose the undesirable chemical substance(s), said apparatus comprising in combination:

(a) a reaction vessel having an interior region to contain an aqueous portion having the undesirable chemical substance(s) during treatment;

(b) an ultraviolet laser device proximate to said reaction vessel capable of generating an ultraviolet laser beam at a wavelength or wavelength range of about 180 nm

to 400 nm (55,560 cm^{-1} to 25,000 cm^{-1}); and,

(c) a laser beam window portion of said reaction vessel that is substantially transparent to ultraviolet laser radiation at wavelengths between about 180 nm to 400 nm (55,560 cm^{-1} to 25,000 cm^{-1}) and is oriented substantially orthogonally relative to the ultraviolet laser beam to pass ultraviolet laser radiation from said ultraviolet laser device into said interior region.

61. (New) Apparatus according to claim 60 further wherein the ultraviolet laser device is capable of producing ultraviolet laser light having at least ten times the energy of conventional ultraviolet lamp light per square millimeter of ultraviolet laser beam cross-section.

62. (New) Apparatus according to claim 60 further comprising an analytical system for continuously monitoring changes in the chemical composition of an aqueous portion in the reaction vessel during irradiation of the aqueous portion with ultraviolet laser radiation from the ultraviolet laser device.

63. (New) Apparatus according to claim 62 wherein said analytical system comprises an interlinked deuterium lamp device, a spectrometer, and a computer system.

64. (New) Apparatus according to claim 63 wherein said deuterium lamp device is oriented to deliver one or more beams of light through a first optically transparent wall portion of the reaction vessel and into the interior region, through the interior region of the reaction vessel including through the aqueous portion therein, out of the interior region through a second optically transparent wall portion of the reaction vessel, and thereafter into the spectrometer.

65. (New) Apparatus according to claim 60 wherein said reaction vessel comprises a quartz tube.

66. (New) Apparatus according to claim 60 wherein the area and shape of said laser beam window portion of said reaction vessel are substantially the same, respectively, as the area and shape of a cross-section of the laser beam generated by the ultraviolet laser device.

67. (New) Apparatus according to claim 60 wherein said ultraviolet laser device is capable of generating a pulsed ultraviolet laser beam capable of delivering an energy density in the range of about 0.10 millijoules to 1 joule per square millimeter per pulse to an aqueous portion in the reaction vessel at a pulse rate of about 1 to 50,000 pulses per second.

68. (New) Apparatus according to claim 60 wherein said ultraviolet laser device generates a monochromatic laser beam at a wavelength of about 193 nm (51,810 cm^{-1}) or lower.

69. (New) Apparatus according to claim 60 wherein said reaction vessel includes fluid inlet and fluid outlet ports such that said aqueous portion having undesirable chemical substances can be continuously flowed through the interior region of said reaction vessel.

70. (New) Apparatus according to claim 69 further comprising valves associated respectively with said fluid inlet and fluid outlet ports for alternately stopping or resuming fluid flow.

71. (New) Apparatus according to claim 69 wherein said fluid inlet and fluid outlet ports are arranged such that the aqueous portion is flowed through said reaction vessel in a direction of flow opposite to the direction of the ultraviolet laser beam through the vessel.

REMARKS

A. The Preliminary Amendment

Claims 31 – 71 are now pending in the present application. Original PCT Claims 1 – 30 have been canceled. Claims 31 – 71 are new claims which have been substituted for original PCT Claims 1 – 30 to more clearly distinguish over the prior art cited in the International Search Report (ISR). No new matter has been added. Each of the new claims is clearly supported by the application as originally filed. On request by the Examiner, Applicants agree to promptly identify the support in the Specification for any new claim or portion thereof.

Entry of this Preliminary Amendment is respectfully requested. It is further requested that the filing fee calculation for this application be based on the new set of claims 31 – 71 submitted herein.

B. Adding Alternative Units of Measurement

The original Specification and Claims defined the UV laser light wavelengths of this invention is "nm" units. To better distinguish over the prior art cited in the International Search Report (ISR), this Preliminary Amendment amends the Specification and claims to also recite the UV laser light wavelengths of this invention in the alternative units of measurement "cm^{-1}." Because this represents a direct and familiar conversion of one unit of measurement to the mathematically equivalent alternative unit of measurement, no new matter has been added by this amendment.

C. The Claims Clearly Distinguish Over the ISR References

The International Search Report, Written Opinion and Preliminary Examination Report in the corresponding PCT application cited and applied three U.S. patent references: U.S. Pat. Nos. 3,941,670 (Pratt '670); 5,120,450 (Stanley '450); and 5,144,146 (Wekhof '146). The claims now pending in this application clearly distinguish over these three references, whether viewed individually or in combination.

1. Pratt '670

Pratt '670 was cited for its teaching of a method of altering (i.e., deactivating or activating) the biological activity of macromolecular species by employing "laser beam radiation at a frequency that excites vibrational and rotational states of the irradiated species...." (see Abstract of Pratt '670). The range of useful laser beam radiation frequencies taught by Pratt '670 is 350 cm^{-1} to 3500 cm^{-1} (see, e.g., Claim 6 of Pratt '670). This range was not selected arbitrarily: it is based on the "Raman spectra of many amino acids, nucleic acids, and biopolymers...." (col. 1, lines 45-49 of Pratt '670).

In contrast to the present invention, however, Pratt '670 teaches nothing whatsoever about decomposing chemical compounds, whether organic or inorganic. Pratt '670 talks about "activating" or "deactivating" certain types of organic molecules, not about destroying those compounds. Furthermore, Pratt '670 uses an irradiation range that is generally regarded as being in the infrared portion of the light spectrum, far from the ultraviolet laser light used by the present invention. Indeed, there is an entire order of magnitude difference between the light frequency range of Pratt '670 (350 cm^{-1} to 3500 cm^{-1}) and the UV laser light range (55,560 cm^{-1} to 25,000 cm^{-1}) taught and claimed in this application.

2. Stanley '450

Stanley '450 was cited for its teaching of a fluid decontamination apparatus that utilizes a combination of high intensity, directed ultraviolet radiation and oxidation. Stanley '450 teaches using ultraviolet light at a particular wavelength of 254 nm, and goes to great lengths to produce UV light at this precise wavelength (see, e.g., col. 4, lines 44-59 of Stanley '450).

Even with UV light at 254 nm, however, Stanley '450 teaches that "the presence of an oxidant" is still required "for promoting the oxidation of organic contaminates" (col. 4, lines 59-62). The oxidants taught by Stanley '450 are hydrogen peroxide and ozone (col. 3, lines 63-65). As taught by subparagraph (d) of Claim 8 of Stanley '450, the presence of an oxidant is necessary for operation of this system because the oxidant creates hydroxyl radicals "resulting in organic molecular disassociation."

By contrast, the methods and apparatus of the present invention do not require the addition of an oxidant to the aqueous solution being treated for removal of contaminants. This leads to important process and reagent efficiencies, added process/apparatus simplicity and flexibility, and resultant cost savings. Nothing in Stanley '450 suggests that there is any way to effectively carry out the Stanley '450 process without addition of an oxidant. Furthermore, Stanley '450 also fails to teach or suggest the particular energy densities, pulse rates, and other important process parameters as recited in the present claims.

3. Wekhof '146

Wekhof '146 was cited for its teaching of a method for destruction of toxic compounds through direct ultraviolet irradiation. Although Wekhof '146 teaches using UV light in a wavelength range of 175-300 nm, it is clear that Wekhof '146 uses UV lamp light,

not UV laser light (see, e.g., col. 3, line 62). Thus, the average power density in Wekhof '145 is maintained at a value "of about 0.1 Watt/cm^2" (col. 4, lines 7-8), or "0.5 Watt/cm^2" (Abstract), both of which are at least an order of magnitude smaller than the power densities used in the present invention.

In addition, none of the three ISR references teach anything about the claimed methods for monitoring the progress of the contaminant decomposition process according to this invention. Also, none of the three ISR references teach or suggest treatment methods that result in substantially complete decomposition of contaminating substances in an aqueous solution in the very short treatment times of this invention; and, there is no basis for anyone to believe that such results are even possible apart from the teachings of this invention.

SUMMARY AND CONCLUSIONS

For all of the foregoing reasons, Applicants respectfully submit that Claims 31 – 71 now pending are patentable over the prior art of record, and an early Notice of allowance is earnestly requested.

Respectfully submitted,

Date Aug. 9, 2006

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Practitioner's Docket No. WLI-001A

CHAPTER II

Preliminary Classification:

Proposed Class:

Subclass:

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INFORMATION DISCLOSURE STATEMENT
ACCOMPANYING NEW APPLICATION TRANSMITTAL

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INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/US2005/009500	22 March 2005 (22.03.2005)	25 March 2004 (25.03.2004)

TITLE OF INVENTION

Ultraviolet Laser System for Decomposing Chemical Pollutants

APPLICANT(S)

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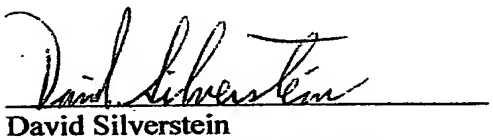
Docket No. WLI-001A

INFORMATION DISCLOSURE STATEMENT

Attached is form PTO-1449A.

Each of the references listed on PTO-1449A was cited in the International Search Report in the PCT application upon which the present Sec. 371 national stage application is based.

Respectfully submitted,



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(Use as many sheets as necessary)

Sheet 1 of 1

Complete if Known	
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First Named Inventor	Waheed Mukaddam
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U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US- 5,120,450	06-09-1992	E. Glynn Stanley, Jr.	
		US- 3,941,670	03-02-1976	George W. Pratt, Jr.	
		US- 5,144,146	09-01-1992	Alexander Wekhof	
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FOREIGN PATENT DOCUMENTS

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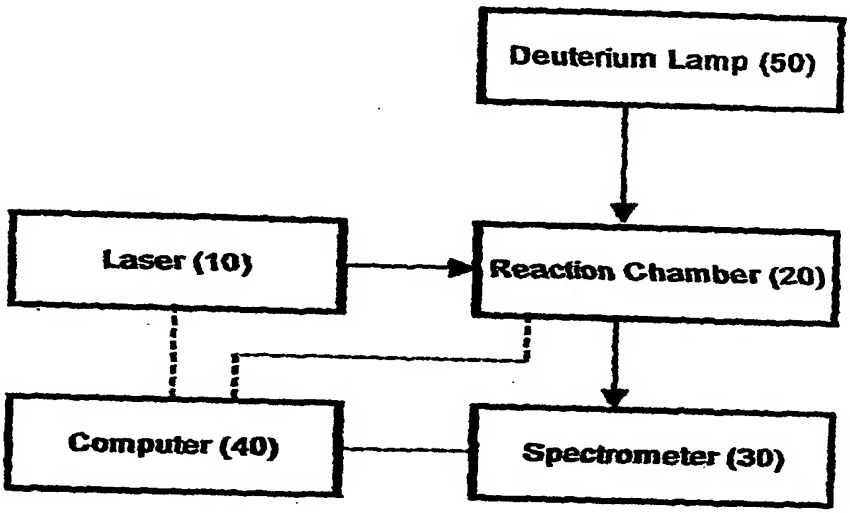

(57) Abstract: Apparatus and methods are disclosed for using an ultraviolet laser system to decompose selected chemical substances in water. More particularly, this invention provides methods and apparatus whereby various environmental pollutants in water are rapidly decomposed to very low concentrations, consistent with environmental discharge regulations, use requirements, and/or applicable health standards, by means of exposure to ultraviolet laser irradiation (10, 20), either with or without one or more catalysts and/or other chemical additives to facilitate or enhance the decomposition process.

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ULTRAVIOLET LASER SYSTEM FOR DECOMPOSING CHEMICAL POLLUTANTS

FIELD OF THE INVENTION

This invention relates generally to apparatus and methods wherein an ultraviolet laser system is used to decompose or otherwise alter the chemical structure of a variety of inorganic and organic chemicals, particularly chemical constituents regarded as pollutants, for example organic contaminants such as fluorinated organic compounds, such as perfluorooctanoate, and inorganic contaminants such as perchlorate, in aqueous solution and/or dispersion.

BACKGROUND OF THE INVENTION

There are a variety of inorganic and organic, partially or fully water soluble, chemical compounds that often appear in wastewater, groundwater and drinking water, which substances are regarded as being of "environmental concern" even at very low concentrations, such as in the 1-10 parts per billion range. Examples of such substances are organic chemicals, such as polychlorinated bi-phenyls, dioxins, 1– 4 dioxane, pentachlorophenol, organic perchlorates, chlorinated solvents, tri- and di-nitro toluene and fluorinated organic compounds such as perfluorooctanoate; and, inorganic chemicals such as inorganic perchlorates. Traditional water purification treatments for these chemical pollutants have included activated carbon adsorption, ultraviolet (lamp) catalyzed peroxide treatment, in-situ biological treatment, membrane filtration and containment, among others. However, the physical, chemical and/or biological properties of these chemical pollutants typically makes recovery, removal and/or decomposition to concentrations below a threshold

concentration of concern prior to discharge to the environment or use of the water containing them very difficult, very expensive, and in some cases impossible.

It is generally known in the art to utilize light energy, typically in the form of an ultraviolet (UV) light source (e.g., for UV light, a mercury vapor lamp) to facilitate or catalyze certain types of chemical or biological reactions. Such processes typically deliver continuous, low-level lamp-generated radiation in the UV range over extended periods of time.

For example, representative of the prior art in this field, U.S. Pat. No. 4,012,321 teaches oxidizing refractory organic compounds in aqueous waste streams by adding hydrogen peroxide and irradiating with UV lamp light.

U.S. Pat. No. 4,661,264 teaches disinfection of water or other fluid by passing a stream of the fluid through a gas pulsed UV laser. Ultraviolet light is taught to be a known disinfection agent for water that kills bacteria by direct contact rather than by secondary photochemical effects.

U.S. Pat. No. 5,877,392 teaches decomposing nonhydrolyzable ambients, e.g., chlorofluorocarbons and nitrogen trifluoride, using UV light and a mediating species (Si) to allow chemical reactions to occur to produce an effluent which can be hydrolyzed by conventional methods. This method can be carried out in gases or liquids.

U.S. Pat. No. 5,178,772 teaches decomposing metal complexes in aqueous solution by adding an oxidizing agent and exposing the solution to UV radiation.

U.S. Pat. No. 6,692,694 teaches a method of deactivating chemical contaminants and biological agents on a surface by aerosol spraying of the surface with an electrostatically charged, photosensitizer solution, followed by illumination with UV light. This patent further teaches, however, that UV light has "limited effect on destroying chemical contamination."

U.S. Pat. No. 4,400,270 teaches a UV sterilization and disinfection system for fluids, such as water for commercial or residential applications, to insure destruction of selected organisms.

U.S. Pat. No. 5,141,636 teaches prolonging the life of a GAC water treatment device by using UV radiation to prevent microbial proliferation on carbon surfaces by oxidizing organic contaminants in the water and thereby disinfecting the water.

Japanese patent publication JP 2001231881 teaches a method and device for decomposing pollutants, such as PCB, furan, and dioxins, to make them non-noxious by irradiation with infrared laser beams.

International patent publication WO 02/45756 teaches a process for oxidizing dangerous chemical and biological materials by spraying a region containing such materials with a gas/vapor cloud of a liquid solution containing a photocatalytic oxidizing substance, followed by directing a high intensity beam of light (wavelength 220-390 nm) across the cloud to trigger a catalyzed activation that releases free radicals that react with and thereby destroy the dangerous chemical or biological materials.

European patent publication EP 0798270 teaches a laser device for purifying contaminated water by destroying bacteriological microorganisms.

U.S. Pat. No. 5,376,281 teaches an apparatus for purifying water using UV radiation to kill and remove microbes.

British patent publication GB 2316528 teaches a process for cleaning or decontaminating the surface of an object (such as a vessel) using a UV laser beam.

Japanese patent publication JP 10075991 teaches cleaning and/or decontaminating metal or polymer surfaces to remove contaminants such as nuclear contaminants by irradiating the surface with a UV laser.

U.S. Pat. No. 6,531,065 teaches a method of treating water to remove perchlorate ion using chemical treatment in combination with UV light from a low pressure mercury lamp (mostly at about 254 nm) to convert perchlorate to a less toxic product.

U.S. Pat. No. 5,258,124 teaches a process for treating aqueous waste water or groundwater containing certain types of organic contaminants using a chemical agent in combination with UV light having a wavelength in the range of about 180-260 nm.

U.S. Pat. No. 6,773,683 teaches a photocatalytic reactor system consisting of a photonic energy source to remove undesirable contaminants (particularly sulfur oxides) from flue effluents to reduce acid rain. In one embodiment, the photonic energy source is a laser.

The foregoing prior art (which is incorporated herein by reference), however, fails to teach a quick, reliable, relatively inexpensive and substantially complete method for decomposing or otherwise chemically altering chemical substances, particularly those regarded as contaminants, in aqueous solutions and/or dispersions prior to use or discharge to the environment. Some of the known water treatment approaches, as referenced above, require treatment and/or UV lamp exposure times that can take many hours. Even with lengthy treatment times, known water treatment approaches may only eliminate about 60-80% of the concentration of chemical contaminants. Other known water treatment approaches may require adding chemicals which themselves can constitute contaminants, or else which react with the contaminant in the water to create environmentally undesirable byproducts.

These and other deficiencies in or limitations of the prior art in this field are overcome in whole or at least in part by the ultraviolet laser system and methods of this invention for treating water and decomposing chemical pollutants therein.

OBJECTS OF THE INVENTION

A general object of the present invention is to provide apparatus and methods for cost-effectively decomposing various chemical substances, particularly pollutants, in aqueous solution and/or dispersion to concentrations that are acceptable for use of the treated water or discharge to the environment consistent with prevailing local, state, federal and/or other environmental regulations or pertinent health standards.

A more specific object of this invention is to provide an ultraviolet light laser system, and methods of using the laser system, to treat water to substantially eliminate selected organic and/or inorganic chemical constituents.

Another object of this invention is to provide a UV-laser based water treatment system and methods that decompose at least selected chemical constituents in very short times, e.g., within about 15 minutes or less, and in some instances as little as a fraction of a second, for example about 0.10 – 1.0 seconds, of treatment time.

Yet another object of this invention is to provide a UV-laser based water treatment system and methods that reliably decompose substantially all, e.g., greater than about 90%, of the undesirable chemical constituents in very short treatment times.

Still another object of this invention is to provide a UV-laser based water treatment system and methods to irradiate water with monochromatic laser light, all or a predominant portion of which is at wavelengths within the ultraviolet range of about 180 nm to 400 nm and, in some preferred embodiments for selected applications, about 193 nm or lower.

Yet a further object of this invention is to provide a UV-laser based water treatment system and methods capable of delivering UV laser light energy that is one or more orders of magnitude (i.e., at least ten times) greater than the light energy provided by conventional UV lamp light to a water sample to be treated.

Still a further object of this invention is to provide a pulsing technique for delivering short bursts of very high energy intensity UV laser radiation of a suitable wavelength or within a suitable wavelength range to a static or flowing water sample to be treated to rapidly decompose selected chemical substances dissolved and/or dispersed in the sample.

These and other objects, advantages and benefits of this invention will be better understood by the following description read in conjunction with Figs. 1 and 2.

SUMMARY OF THE INVENTION

It has been found in accordance with this invention that irradiation of water solutions and/or dispersions of certain classes of chemical substances by means of an ultraviolet laser, in some cases with or in some cases without suitable catalysts, will effectively and efficiently result in the substantially complete decomposition of these chemicals in relatively short treatment times, typically about 15 minutes or less, preferably less than about 10 minutes, more preferably less than about 5 minutes, even more preferably less than about one minute, and, in some instances, less than one second, for example about 0.10 – 1.0 seconds. A process in accordance with this invention utilizing an ultraviolet laser to decompose environmentally refractory chemicals in water results in a cost-effective method of realizing pollutant concentrations that are below threshold concentrations of environmental concern such that the treated water can then be safely used or discharged to the environment.

More particularly, it has been found in accordance with this invention that the very high energy intensity of laser-generated ultraviolet light pulses in combination with the monochromaticity of UV laser light has a surprising and dramatic impact on at least certain types of undesirable chemical substances that may commonly be present in an aqueous solution and/or dispersion. Such chemical substances are substantially completely decomposed (e.g., at levels of about 90% or higher decomposition) typically in a matter of

only a few minutes or less of exposure of a water sample containing the chemicals to UV laser radiation, preferably at one, or several, or within a range of monochromatic UV wavelengths of about 180 nm to 400 nm, in some instances preferably about 193 nm or lower. It has been consistently found that a UV laser treatment in accordance with this invention results in higher decomposition/destruction of chemical constituents in a treated aqueous sample over a dramatically shorter period of treatment time than does light from other photonic sources, specifically light from a UV lamp.

In a preferred embodiment of this invention, the UV laser radiation wavelength(s) used is (are) selected in relation to the chemical substance(s) to be decomposed in a particular aqueous sample. It has been found that UV laser radiation wavelengths or wavelength ranges can be effectively coordinated with chemical atomic bonding energies. Thus, in accordance with an embodiment of this invention, it may be possible to identify a preferred UV laser radiation wavelength for decomposing a single chemical constituent, or to identify preferred multiple UV laser radiation wavelengths, or a preferred UV laser radiation wavelength range, for decomposing multiple chemical constituents or for addressing a single chemical constituent that may decompose into different intermediate substances prior to complete decomposition. In some instances, it has been found that UV laser radiation at about 193 nm or lower, i.e., about 180 nm – 193 nm, is particularly effective in decomposing many of the chemical constituents of primary interest.

Chemicals which have been found to be effectively decomposed by the UV laser treatment of this invention include inorganic chemicals such as inorganic perchlorates, and organic chemicals such as polychlorinated bi-phenyls, dioxins, 1-4 dioxane, pentachlorophenol, tri- and di-nitro toluene, chlorinated organic solvents, and fluoriated organic compounds such as perfluorooctanoate. A particular group of chemicals that has proven to be especially responsive to treatment in accordance with this invention are the

polyfluorinated straight chain organic acids having eight or more carbon atoms. The foregoing list of chemicals which may be effectively decomposed in aqueous solution and/or dispersion according to the practice of this invention is not complete. It will be a matter of routine experimentation by one of ordinary skill in this art to identify other chemically or structurally similar chemicals that respond equally well to the laser decomposition treatment of this invention. All of such chemicals are intended to be encompassed by the present invention.

The present invention thus relies on a combination of the highly monochromatic properties of laser light, on the high density of light energy that can be delivered to an aqueous sample by laser radiation, and, in some embodiments, on the selection of a UV laser light wavelength or wavelength range selected relative to the nature and strength of the chemical bonds that need to be broken to effectively decompose a chemical or chemicals in an aqueous solution or suspension. More particularly, it has been determined in accordance with preferred embodiments of this invention that a UV laser can be used to treat a water sample with a laser light energy density of at least about 0.01 millijoules (mJ) per square millimeter per pulse, and perhaps even lower, to as high as 1.0 joule per square millimeter per pulse, or even higher as improvements in laser technology may permit. Based on existing laser technology, a useful laser energy density range for the practice of this invention is about 0.10 to 10 millijoules per square millimeter per pulse, and in some specific tests performed about 0.25 to 2.0 millijoules per square millimeter per pulse. UV laser treatment of water samples within such energy/unit area ranges has been found effective in rapidly decomposing the chemical constituents of interest.

In another preferred embodiment of this invention, the UV laser light is delivered to a water sample containing a chemical to be decomposed in a series of short bursts or pulses. Such pulsed delivery of the UV laser light is limited to some extent by existing laser

technology but may, in general, range from a pulse rate of about 1 pulse/second (or perhaps less in some instances) to about 50,000 pulses/second (50K hertz) or even higher as evolving laser technology permits. A preferred pulse rate range based on existing technology might be from about 10 – 1000 pulses per second. In some specific tests performed, a pulse rate of about 25 – 100 pulses/second has been used effectively. It is also within the scope of this invention to gradually ramp up and/or ramp down the pulse rate to or from the preferred pulse rate.

It should be apparent to those of ordinary skill in this art that various combinations of laser pulse rates and UV laser energy densities are capable of delivering substantially the same quantity of total UV laser energy to a water sample over any given treatment time period. Within reasonable limits, it is believed that any combinations of laser pulse rate and UV laser energy density that deliver substantially the same total UV laser energy to a water sample over a given treatment time will achieve a substantially similar degree of decomposition of chemical constituents in the treated water sample. It would therefore be a matter of routine experimentation to select or determine a suitable combination of laser pulse rate and UV laser energy density for a particular application.

In some embodiments of the present invention, the UV laser chemical decomposition process of this invention can be enhanced by adding relatively small but effective catalytic amounts of one or more non-environmentally sensitive catalysts to the water containing the chemical(s) to be decomposed. Such catalysts advantageously include, for example, iron compounds and hydrogen peroxide. It is hypothesized that some of the UV laser treatment chemical decompositions in accordance with this invention may occur primarily by a UV laser-activated hydrolysis reaction, whereas other UV laser treatment chemical decompositions operate primarily by means of a UV laser-activated reduction reaction. Because the latter reduction reaction is promoted by the presence of an electron acceptor,

such decomposition treatments may especially benefit from the presence of a suitable catalyst. It may be that some chemical decompositions in accordance with this invention operate in part by both of these mechanisms, or, perhaps, by other mechanisms that have not yet been identified.

In other embodiments, the present invention may be practiced to treat water in a batch, semi-batch or continuous process. In a batch process, for example, a water sample to be treated in accordance with the present invention might be placed in an open or closed container and exposed to suitable UV laser light of appropriate wavelength for an effective period of time and at an energy level sufficient substantially to decompose the chemical(s) in the water. Mixing or agitation of the aqueous sample during treatment could optionally be used to assure that all of the water in the container is exposed to the UV laser light.

Alternatively, in a continuous treatment process, water containing a chemical or chemicals to be decomposed could be exposed to UV laser light while flowing through a conduit. In one such embodiment, at least a portion of the conduit wall could be substantially transparent to UV laser light of the appropriate wavelength. For example, the conduit might include one or more optical "windows" along its length fashioned, for example, from quartz. At each such optical window, suitable UV laser light would be directed through the window into the flowing stream of water. The flow rate of the water through the conduit, or the number of optical windows/laser stations, or both could be varied as needed to assure that the water receives exposure to the UV laser light for an effective period of time sufficient to substantially decompose the chemical(s) in the water.

In an alternative embodiment of such a continuous flow treatment process, the entire conduit, or at least a treatment portion of a conduit, could be made of quartz so as to be transparent to UV laser light along its entire length. Multiple laser beams could be used to deliver laser radiation to an aqueous stream flowing through such a conduit either at one or

more discrete locations along the length of the conduit or substantially continuously along the length, or at least along a portion of the length, of the conduit.

In still another embodiment of a continuous flow treatment process in accordance with this invention, as schematically illustrated in Fig. 2, a generally linear conduit or conduit portion 20 may be provided with a fluid inlet port 22 at a first end of a conduit portion, a fluid outlet port 24 at a second end of a conduit portion, and a UV laser radiation-optically transparent window 26 (made, for example, of quartz) at at least one or the other conduit end, with the plane of the window positioned substantially perpendicular to the longitudinal axis of the conduit. In a preferred embodiment of this practice of the invention, the shape and size of the optical window is matched to the shape and size of the conduit and to the shape and size of the UV laser beam (or vice versa) in order to fully utilize all of the UV laser light and to irradiate aqueous sample even along the inner wall of the conduit.

Using this apparatus configuration, UV laser radiation can be directed through the optical window 26 so as to pass through fluid contained or flowing in the conduit with the laser radiation running substantially parallel to the longitudinal axis of the conduit 20. In this apparatus configuration, the direction of fluid flow through the conduit may be either "co-current" (in the same direction as the direction of the UV laser beam) or "counter-current" (in the opposite direction as the direction of the UV laser beam).

Any of the aforementioned and described continuous flow treatment processes of this invention may also be adapted to semi-batch processes. In such a semi-batch process, an aqueous sample to be treated would be flowed into a treatment portion of a conduit. Fluid flow would be temporarily stopped for a period of time, such as by closing valves, such as valves 23 and 25 as seen in Fig. 2, associated with the respective fluid inlet and fluid outlet ports, while the sample in the treatment conduit was treated with UV laser light in accordance with this invention. The period of time for stopping fluid flow would ordinarily be less than

about 15 minutes, as described above in accordance with other invention embodiments. When the treatment had resulted in substantially complete decomposition of the subject chemical constituents, as determined for example by suitable continuous or periodic monitoring means, the treated aqueous sample would be flowed out of the treatment portion of the conduit, for example by opening valves associated with the respective fluid inlet and fluid outlet ports, and a new aqueous sample flowed into the treatment conduit.

In another embodiment of this invention, as illustrated in Fig. 1, previously-described treatment embodiments of this invention may be advantageously coupled with an aqueous sample composition continuous or semi-continuous monitoring process in conjunction with an associated monitoring system. Thus, at the same time that an aqueous sample (flowing or static) is being treated with UV laser radiation in accordance with this invention, it may be useful to continuously monitor the chemical composition of the aqueous sample, for example to determine when the decomposition of chemical constituents is substantially completed. It is known generally in the chemical arts to locate various types of sensors (e.g., temperature sensors) in a fluid undergoing chemical changes, or, alternatively, to periodically withdraw fluid samples, to monitor the rate and progress of such chemical changes, and such conventional approaches are within the scope of the present invention.

In a preferred invention embodiment, however, as schematically illustrated in Figs. 1 and 2, at the same time that a UV laser beam 12 from a laser unit 10 is being passed through an aqueous sample in a first direction, another (second) light beam 52 (such as from a deuterium lamp) is passed through the aqueous sample being treated, at one or a plurality of locations, in a second direction that is preferably orthogonal to the first (UV laser beam) direction. Because light beams do not interfere with one another, these two steps (UV laser treatment and spectrophotometric monitoring) can be carried out simultaneously. Such a step can readily be carried out if the aqueous sample is in a treatment vessel or conduit made

completely of a light-transparent material such as quartz or in a conduit having optical windows located along its wall portions. By monitoring the deuterium lamp light 54 (such as with a spectrometer 30) emerging from the aqueous sample that is being treated with the UV laser, the progress of the decomposition of chemical constituents in the aqueous sample can be continuously monitored. When the rate of change of the emerging deuterium lamp light substantially levels off, it can be concluded that chemical decomposition in the aqueous sample is substantially complete.

Other ways of practicing this invention will be apparent to those of ordinary skill in this art, and such alternative embodiments of this invention are intended to be covered by this application.

BRIEF DESCRIPTION OF THE DRAWING

Fig. 1 is a schematic process flow diagram illustrating one embodiment for carrying out an ultraviolet laser decomposition and monitoring process in accordance with this invention.

Fig. 2 is a schematic illustration of a continuous flow treatment embodiment of the present invention in combination with a chemical composition monitoring system according to this invention.

DETAILED DESCRIPTION OF AN EMBODIMENT OF THE INVENTION

A laboratory test system for carrying out an embodiment of the present invention is illustrated in the block flow diagram of Fig. 1. As shown in Fig. 1, the principal apparatus/ process elements consist of an ultraviolet (UV) laser 10, a reaction vessel, or conduit or region 20 to contain a sample of water and chemical substance(s) therein, a deuterium lamp 50, an ultraviolet spectrometer 30 and a computer system 40. A laser beam from the UV

laser 10 is directed at the reaction vessel 20 through a substantially UV-transparent optical window portion (identified by reference numeral 26 in Fig. 2) of vessel 20 so as to irradiate the contents of the reaction vessel. The deuterium lamp, which is part of an ultraviolet spectrophotometer system, is preferably oriented so as to produce light beams 52 substantially at right angles to the laser beam 12 and provides a means of continuously monitoring the ultraviolet spectrophotometric signature of the contents of the reaction vessel and thus provides an indication of the chemical reactions that are occurring within the reaction vessel. Water containing a known concentration of the chemical(s) of interest is placed in the reaction vessel in accordance with this invention; and, the ultraviolet spectrophotometric signature and temperature of the contents of the reaction vessel are recorded before the laser is activated. Thereafter, the laser is activated, and the ultraviolet spectrophotometric signature and temperature of the contents of the reaction vessel are continuously measured and recorded. After a period of laser irradiation, usually less than about 15 minutes, for example an irradiation period of from about 1 second to about 10 minutes, in some embodiments from about 10 seconds to about 5 minutes, in other embodiments less than one second, the ultraviolet spectrophotometric signature of the sample has significantly changed indicating substantial decomposition of the chemical substance(s) and the concurrent detoxification of the water. The laser is turned off when the rate of change of the spectrophotometric signature slows or ceases indicating substantial completion of the chemical decomposition process. The irradiated sample is then removed from the reaction vessel for further analysis.

An irradiated sample can be analyzed, for example by means of FTIR analysis, to identify the decomposition products, and also by wet chemistry to determine the residual concentration(s) of the chemical substances remaining in the sample after the radiation step. The concentration(s) of the chemical substances in the irradiated sample is compared to the

concentration(s) in a sample that has not been irradiated to judge the extent (success) of the decomposition process. Samples of varying concentrations of pollutants can be irradiated as described above, and data for these samples can be recorded and compared. Catalysts such as iron and chemicals such as hydrogen peroxide can be optionally utilized to facilitate, enhance, and/or expedite the UV laser-promoted chemical decomposition process of this invention.

Example

For example, in demonstrating the practice of one embodiment of this invention, a distilled water blank and various concentrations, ranging from about 5 ppm to about 500 ppm, of ammonium perfluorooctanoate (apfo) in distilled water were prepared. A portion of each apfo sample was set aside as a reference and another portion of each sample was introduced into a 22mm diameter by 50mm long synthetic quartz (Suprasil™) reaction vessel. The water solutions of apfo were irradiated by means of an excimer laser (LambdaPhysik LPX210i) producing a monochromatic 193nm wavelength beam operating at an energy level of 100 millijoules per pulse at a frequency of 50 pulses per second. The laser beam was directed along (parallel to) the long axis of the reaction vessel and completely covered an 8mm high by 22mm wide optical window section of the reaction vessel so as to fill an 8mm by 22 mm portion of the reactor cavity with UV laser light resulting in the delivery of a UV light energy intensity of about 0.57 millijoules/square millimeter/pulse to the sample. A 30w deuterium lamp and Zeiss MMS-UV spectrometer were mounted perpendicular to the long axis of the reaction vessel and approximately in the middle of the reaction vessel to measure the ultraviolet spectrum of the contents of the reaction vessel as a function of time while the sample was being exposed to the UV laser light. Various parameters from the experimental system were interfaced to a computer system electrically

connected to the reaction vessel and the spectrometer. Samples were exposed to the UV laser beam until there ceased to be any appreciable further changes in the spectrophotometric signature of the sample (typically less than about 15 minutes). After exposure to the UV laser beam, the samples and reference solutions were sent to laboratories for FTIR and chemical analyses.

The following is a summary of the testing results:

1. The laser beam passed through the 50 mm (long axis) column of distilled water blank in the reaction vessel essentially unimpeded, and the UV spectrophotometric signature of water did not change as a result of the UV laser irradiation.
2. The laser beam was substantially absorbed in the 50 mm column of apfo - water solution at any of the concentrations tested.
3. Over a period of less than 15 minutes of irradiation, all apfo samples showed significant changes in their UV spectrophotometric signatures.
4. The rate of change of the UV signatures of apfo samples varied over the period of irradiation.
5. The temperature of the apfo samples increased from room temperature to approximately 45°C over the duration of each of the tests runs.
6. Small bubbles formed at the laser input face of the reaction vessel, presumably as a result of the UV laser irradiation treatment. The quantity of the generated bubbles appeared to be directly proportional to the initial concentrations of apfo in the respective samples.
7. FTIR analysis of the irradiated and un-irradiated samples indicated that, in all cases, there were distinct and substantial changes in the FTIR spectra after irradiation with the UV laser.
8. Quantitative analysis of the irradiated samples confirmed greater than 95% decomposition of the apfo.

The following is a summary discussion of the results presented above:

1. Distilled water in the reaction chamber was transparent to the laser beam.
2. Apfo contained in the samples tested absorbed UV light from the UV laser irradiation.
3. Due to the dramatic change in the UV spectrophotometric signatures of the samples during the irradiation treatment, it is concluded that significant chemical changes occur related to the decomposition of apfo.
4. Variations in the rate of the change of the UV spectrophotometric signatures of the irradiated samples during treatment strongly suggest the formation of some numbers of intermediate species, which are subsequently decomposed by further treatment with UV laser radiation in accordance with this invention.
5. The increase in the temperature of the apfo samples during irradiation suggests that photon energy is being transferred from the UV laser beam and converted, at least in part, into thermal energy; the increase in sample temperature during irradiation may also in part be the result of heat released from chemical reactions promoted by the UV laser irradiation.
6. Because gas bubbles did not appear during the irradiation of the distilled water sample, and the quantity of the gas bubbles appeared to be proportional to the initial apfo concentration in the samples, the gas bubbles are thought to be a gaseous reaction product from the laser decomposition of apfo in accordance with this invention.
7. FTIR analysis results showed shifts in both spectral peaks and heights that would be consistent with apfo decomposition. The FTIR signatures of the irradiated samples indicated decomposition of the larger molecules into two-carbon molecules.
8. Quantitative analysis of the irradiated samples confirmed greater than 95% decomposition of the apfo.

It will be apparent to those skilled in the art that changes and modifications may be made in the above-described apparatus and methods for an ultraviolet laser system for decomposing chemical substances in water without departing from the spirit and scope of the invention herein, and it is intended that all matter contained in the above description shall be interpreted in an illustrative and not a limiting sense.

Having described the invention, what is claimed is:

CLAIMS

1. A method for substantially completely decomposing selected chemical substance(s) in an aqueous solution and/or dispersion, said method comprising the step of exposing an aqueous portion containing a chemical substance(s) to ultraviolet laser irradiation at a suitable wavelength and of sufficient energy for a sufficient period of time substantially to decompose the chemical substance(s) in the aqueous portion.

2. A method according to claim 1 wherein said ultraviolet laser irradiation has a wavelength of about 180 nm to 400 nm and delivers an energy density in the range of about 0.10 to 10 millijoules per square millimeter to said aqueous portion.

3. A method according to claim 1 wherein said ultraviolet laser irradiation has a wavelength in the range of about 180 nm to 193 nm.

4. A method according to claim 1 wherein the total period of time during which the aqueous portion is exposed to the ultraviolet laser irradiation is about 1 second to 10 minutes.

5. A method according to claim 1 wherein the total period of time during which the aqueous portion is exposed to the ultraviolet laser irradiation is less than one second.

6. A method according to claim 1 wherein 90% or more of at least one chemical substance originally present in the aqueous portion is decomposed within an ultraviolet laser irradiation period of less than 15 minutes.

7. A method according to claim 1 wherein the aqueous portion before treatment contains one or more selected chemical substances selected from the group consisting of perchlorates, polychlorinated bi-phenyls, dioxins, 1-4 dioxane, pentachlorophenol, tri- and di-nitro toluene, chlorinated solvents, fluorinated organic compounds, and mixtures thereof.

8. A method according to claim 1 wherein the aqueous portion before treatment contains at least a polyfluorinated straight chain organic acid having at least eight carbon atoms.

9. A method according to claim 1 wherein a catalyst is added to the aqueous portion before or during the ultraviolet laser irradiation step.

10. A method according to claim 1 wherein said ultraviolet laser irradiation is delivered to the aqueous portion in pulses at a pulse rate ranging from about 1 to 50,000 pulses per second.

11. A method according to claim 1 wherein said ultraviolet laser irradiation is delivered to the aqueous portion in pulses at a pulse rate ranging from about 10 to 1000 pulses per second.

12. A method according to claim 1 wherein said ultraviolet laser irradiation is delivered to the aqueous portion in pulses at a pulse rate ranging from about 25 to 100 pulses per second.

13. A method according to claim 10 further comprising the step of selecting a combination of ultraviolet laser pulse rate and energy density so as to deliver to the aqueous portion sufficient laser energy to effect substantially complete decomposition of said chemical substance(s) within a total treatment time of about 15 minutes or less.

14. A method according to claim 10 further comprising the step of selecting a combination of ultraviolet laser wavelength or wavelengths, ultraviolet laser pulse rate and energy density so as to deliver to the aqueous portion sufficient laser energy to effect substantially complete decomposition of said chemical substance(s) within a total treatment time of about 15 minutes or less.

15. A method according to claim 1 further comprising a monitoring step of periodically or continuously monitoring the chemical composition of the aqueous portion during the ultraviolet laser treatment.

16. A method according to claim 15 wherein said aqueous portion is treated with ultraviolet laser light while flowing in a conduit, and said monitoring step is performed continuously during laser treatment at two or more locations along said conduit.

17. A method according to claim 15 wherein said monitoring step comprises the steps of passing a light beam through an optically transparent wall portion of a container holding said aqueous portion during the ultraviolet laser treatment, through the aqueous portion in said container, out of said container through an optically transparent wall portion,

and into a spectrometer for monitoring the spectrophotometric signature of the aqueous portion during ultraviolet laser treatment.

18. A method according to claim 1 wherein the aqueous portion is exposed to the ultraviolet laser irradiation in a batch, semi-batch, or continuous flow process.

19. Apparatus for treating an aqueous solution and/or dispersion of selected chemical substance(s) so as to substantially completely decompose the chemical substance(s), said apparatus comprising in combination:

(a) a reaction vessel having an interior region to contain an aqueous portion containing the chemical substance(s);

(b) an ultraviolet laser proximate to said reaction vessel for generating an ultraviolet laser beam at a wavelength or wavelength range of about 180 nm to 400 nm; and,

(c) a window portion of said reaction vessel that is substantially transparent to ultraviolet laser radiation at wavelengths between about 180 nm to 400 nm to pass ultraviolet laser radiation from said laser into said interior region.

20. Apparatus according to claim 19 further wherein the ultraviolet laser beam is in alignment with said window portion of the reaction vessel for coupling laser light into the interior of the vessel.

21. Apparatus according to claim 19 further comprising an analytical system for continuously monitoring changes in the chemical composition of the aqueous portion in the reaction vessel during irradiation of the aqueous portion with ultraviolet laser radiation from the ultraviolet laser.

22. Apparatus according to claim 21 wherein said analytical system comprises a deuterium lamp device, a spectrometer, and a computer system.

23. Apparatus according to claim 22 wherein said deuterium lamp device is positioned to deliver one or more beams of light into an optically transparent wall portion of the reaction vessel, through the interior region of the reaction vessel including through the aqueous portion therein, out through an optically transparent wall portion of the reaction vessel, and thereafter into the spectrometer.

24. Apparatus according to claim 19 wherein said reaction vessel comprises a quartz tube.

25. Apparatus according to claim 19 wherein the area and shape of said window portion of said reaction vessel is substantially the same as the area and shape of a cross-section of the laser beam generated by the ultraviolet laser.

26. Apparatus according to claim 19 wherein said ultraviolet laser generates a pulsed laser beam capable of delivering an energy density in the range of about 0.10 millijoules to 1 joule per square millimeter per pulse to the aqueous portion at a pulse rate of about 1 to 50,000 pulses per second.

27. Apparatus according to claim 19 wherein said ultraviolet laser generates a monochromatic laser beam at a wavelength between about 180 nm and 193 nm.

28. Apparatus according to claim 19 wherein said reaction vessel includes fluid inlet and fluid outlet ports such that said aqueous portion can be continuously flowed through the interior region of said reaction vessel.

29. Apparatus according to claim 28 further comprising valves associated respectively with said fluid inlet and outlet ports for alternately stopping or resuming fluid flow.

30. Apparatus according to claim 28 wherein said aqueous portion is flowed through said reaction vessel in a direction of flow opposite to the direction of the ultraviolet laser beam through the vessel.

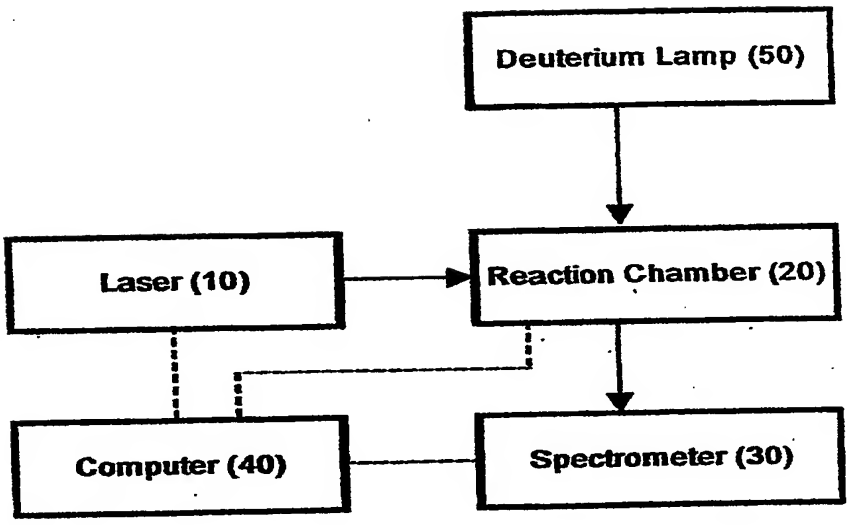

Figure 1

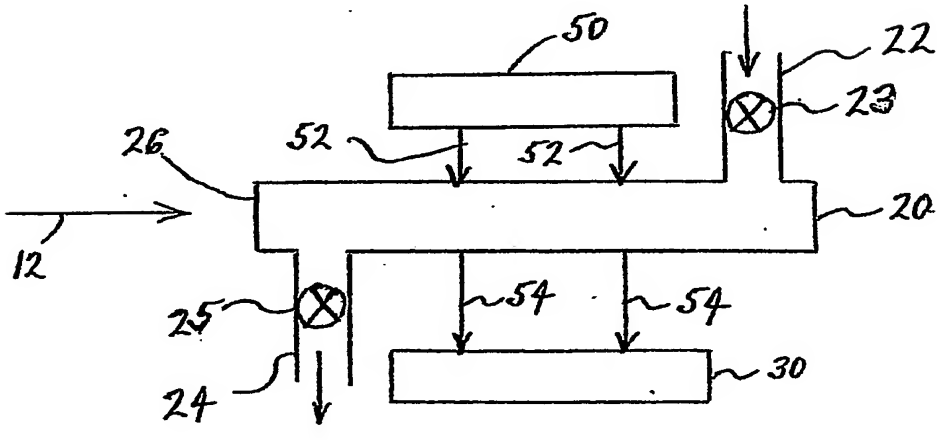

Figure 2

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US05/09500

A. CLASSIFICATION OF SUBJECT MATTER
IPC(7) : A61L 2/08
US CL : 422/24, 22, 186.3; 210/748, 759, 760; 250492.1, 435
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
U.S. : 422/24, 22, 186.3; 210/748, 759, 760; 250492.1, 435

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
Please See Continuation Sheet

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X — Y	US 3,941,670 A (PRATT, JR.) 02 March 1976 (02.03.1976), see entire document.	1-18, 21-25 — 19-20, 26-30
X	US 5,120,450 A (STANLEY, JR.) 09 June 1992 (09.06.1992), see entire document.	1-30
Y	US 5,144,146 A (WEKHOF) 01 September 1992 (01.09.1992), see entire document.	1-30

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:

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Date of the actual completion of the international search
10 June 2005 (10.06.2005)

Date of mailing of the international search report
24 JUN 2005

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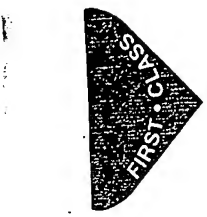
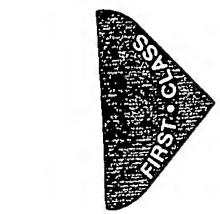



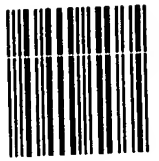

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